RESEARCH IN LASER PROCESSES

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RESEARCH IN LASER PROCESSES

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Calculations have been initiated to determine the threshold conditions and speed for propagation of the previously modeled constricted (arc-like) discharges which often terminate high power, electrical discharge lasers. A "complete" set of electron collision cross sections for CO has been assembled and tested against the meager electron transport data. Measurements of the Tl-Xe fluorescence in the vicinity of the 377.6 and 535 nm lines have been used to calculate the expected gain and absorption of this promising excimer laser candidate. Measurements of integrated absorption, forward (cont. back)

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20. ABSTRACT (cont.) scattering and backward scattering of radiation near the resonance lines have been made over a wide range of sodium densities and will be used to test recent calculation techniques and to determine quenching rate coefficients.

FINAL REPORT

This Final Report contains descriptions of work carried out under ONR Contract No. N0014-67-A-0405-008 and ARPA Order No. 2683, Amd. 1, and covers the period from 1 July 1974 to 30 June 1975. Section I is the Final Report Summary while Sections II-V are more detailed descriptions of work carried out under the four projects supported by this contract.

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I. FINAL REPORT SUMMARY

The four projects being carried out in the area of Laser Processes under this contract are summarized below. More detailed discussions are given in Sects. II through V of this report.

1) Stability of Discharges in Weakly Ionized Gases.

The technical problem being investigated is the optimization of the use of electrical energy for the production of excited molecules in electrically excited gas lasers. In particular, the efficient use of electrical excitation in gas lasers requires that the discharge remain diffuse as the density of the excited molecules is raised as high as possible, i.e., that the discharge not form a constricted channel or arc. During the first six months of this contract we completed numerical calculations of the time dependent electron and ion density, the vibrational and translational temperatures, and the gas density for cylindrically symmetric discharges in a gas mixture typical of those used in high power CO_2 lasers. During the second six months we initiated calculations of the propagation of such discharges in the axial or electric field direction using simplified models in which the gas density and temperature was held constant.

2) Electron Transport and Ionization Coefficients

The objective of this project is to supply accurate electron transport coefficients, electron excitation and ionization coefficients, electron collision cross sections, and excited state relaxation data for use in the electrical discharge models discussed above and being carried out in laser development laboratories. The procedures used include the assessment of the accuracy to be expected from various experimental methods for the measurement of electron transport coefficients; the analysis of published

and unpublished electron transport data and collision cross section data to obtain the best available set of electron cross section data, e.g., a set which is consistent with the limited amount of measured electron transport coefficients in gases of laser interest; and the compilation and application to laser gas mixtures of data on the collisional and radiative destruction of electronically excited molecules. Specific results include the assembly and evaluation of a complete set of electron excitation cross sections for CO, the completion of calculations of the errors in electron drift velocity measurements caused by electron absorption at the electrodes and initial measurements of net rate coefficients for electron excitation of O_2 molecules to the $^1\Sigma_g^+$ state via direct excitation and via dissociative excitation of $O(^1D)$ and subsequent excitation transfer. In accordance with current laser interests we plan to follow this work with measurements of the electron excitation rate coefficients for the $A^3\Sigma_u^+$ state of N_2. Excitation transfer from this metastable state to radiating atoms or molecules is being investigated by a number of laboratories as potential "visible" laser systems.

3) Generation and Interpretation of Molecular Continuum Radiation

Many metal-noble gas molecules are viable candidates for high-efficiency, high-power excimer lasers. Since the spectra and potentials of these molecules are unknown, the merits of different metal-noble gas combinations can only be assessed from detailed measurements of the kind reported here. The alkali-noble gas molecules, which we have studied in detail under this and previous contracts, are candidates for excimer lasers in the 0.7-1 μm wavelength region. Here we report on the Tl-Xe molecules, which could provide excimer laser operation in the 0.43-0.46 μm

(blue) and 0.56-0.7 μm (yellow-red) wavelength regions. From our measurements, we have predicted the absorption and stimulated emission coefficients for Tl-Xe mixtures. Based on these results, a fairly small (2%) fraction of excited Tl (7^2S) is sufficient for useful gain in the above wavelength regions.

Because of the potential of these metal vapor-rare gas systems for laser operation, we have proposed to extend the measurements to systems such as MgXe, which might provide laser action in the 350 nm wavelength region. Also, we plan to determine the contributions of species such as $NaXe_2$ to the continuum fluorescence and net laser gain. Such species may well be important at the high gas densities of proposed laser systems.

4) Scattering and Transport of Resonance Radiation in Gases

The technical problem considered here is the accurate and rapid prediction of the experimentally important loss of excited atoms from laser plasmas through the transport of resonance radiation to the wall of the laser. The method used is the experimental measurement of the magnitude and wavelength dependence of the scattering of radiation incident on a cell filled with metal vapor, e.g., sodium or potassium. The experimentally measured spectral intensity is then compared with theoretical predictions so as to evaluate the accuracy of the various available theories. Experimental measurements of the absorption, backscattering and forward scattering have been made for a wide range of sodium densities and "white light" illumination. We have initiated comparisons of this experimental data with very detailed theories and with approximate theories to test their usefulness for excimer laser discharge models.

II. STABILITY OF DISCHARGES IN WEAKLY IONIZED GASES (Drs. E. F. Jaeger (to 10/74), H.-C. Chen (from 2/75) and A. V. Phelps)

The results of calculations of the time dependent radial distributions of electron, ion and gas densities and of translational and vibrational temperatures in gas mixtures appropriate to high power CO_2 lasers have been discussed in the last Semiannual Technical Report and will not be repeated here. Instead we will discuss the recently initiated calculations of the propagation the constricted discharge in directions parallel to the applied electric field.

The propagation of filamentary electrical discharges in the general direction of the electric field is a commonly observed feature during the electrical breakdown of air across long gaps in the laboratory and in lightning. A few observations of discharge propagation have been made laser systems. These experiments show relatively low propagation velocities ($\sim 10^6$ cm/sec) in the pin-type TEA lasers and higher propagation velocities in the parallel plate or rail type discharges. The general picture of the propagating discharge is that because of the higher drift velocity of the electrons there is a separation of charge which results in a perturbed electric field. If the discharge is of limited extent in the direction perpendicular to the applied field, then field nonuniformities parallel to the applied field will be present and can lead to enhanced ionization. If conditions are right the discharge will propagate as a narrow channel with a high degree of ionization, i.e., an arc channel will form and laser operation will cease. The objective of this project is to combine our previous calculations of radial constriction of the discharge with calculations of the axial propagation which can occur for a radially nonuniform discharge.

Axial non-uniformities in laterally uniform discharges, e.g., cathode fall regions and striations, are not the object of these calculations.

The calculations of the effects of charge separation in the axial direction requires the addition of some form of Poisson's equation to the continuity, momentum and energy equations used previously. In order to simplify the initial calculations and allow more direct comparison with previously published work, we initially assume the gas density and temperature to be independent of position and the electric field and electron and ion densities to vary only in the field direction. In addition, the roles of vibrationally excited states and negative ions are neglected. Two different techniques for calculating the electric field are being investigated, i.e., the conventional Poisson's equation and a form based on Ampere's law. Considerable attention is being given to the formulation of the boundary conditions so as to avoid the development of space charge sheaths at electrodes and the resultant undesired use of computer space. Plans for continuation of this work under the proposed new contract include the modification of the electric field so as to simulate the effects of a finite diameter to the ionized volume, the addition of the effects of negative ion formation and destruction, the change of the electron transport coefficients to those characteristic of high power CO lasers and the addition of the effects of vibrational excitation of the CO.

III. ELECTRON TRANSPORT AND IONIZATION COEFFICIENTS (Drs. S. A. Lawton (from 4/75), J. H. Whealton and A. V. Phelps)

1) In an effort to improve the accuracy of predictions of the power output and efficiency of CO lasers, we have assembled a "complete" set of electron excitation and momentum transfer cross sections based on data which has become available since 1967 [R. D. Hake and A. V. Phelps, Phys. Rev. 158, 70 (1967)]. Unfortunately, the only additional electron transport coefficient data available are measurements of the Townsend ionization coefficient. Thus, there is still no electron drift velocity data for $E/N > 2.5 \times 10^{-16}$ V-cm^2 and the characteristic energy data for $\varepsilon_k > 0.7$ eV is over 50 years old. As a result of this lack of data, the only test we can make of these cross sections for average electron energies above 1 eV is to note that the calculated ionization coefficients are lower than experimental values by a factor of three. Since the electronic excitation cross sections used are largely based on approximate theory, the simplest corrective action would be a reduction of all electronic excitation cross sections by a, as yet undetermined, constant factor. However, it is to be noted that there is approximately a factor of two ratio of calculated to measured vibrational excitation rate coefficients at electron energies between 0.3 and 1 eV when the cross sections measured by electron beam techniques are used for the calculations. Measurements of electron drift velocity and characteristic energy over a wide range of E/N and CO-He mixtures would go a long way toward resolving those uncertainties of importance for the modeling of CO lasers.

2) Since the measurement of electron transport coefficients has proven to be useful and potentially very accurate source of electron rate coefficient

and collision cross section data, it is important that there be reliable evaluations of the sources of error resulting when the measurements are interpreted in terms of the conventional, idealized transport coefficients. Of particular importance in the interpretation of electron drift velocity measurements is the change in the apparent mean transit time caused by the injection and subsequent backscattering of electrons at the cathode of the drift tube.

We have completed an analysis this problem using both the conventional continuity equation for the electron density and the Monte Carlo technique. The results of the Monte Carlo calculation are illustrated in Fig. 1. These calculations have been carried out for a ratio of energy relaxation distance to drift tube length β of 1/30, a ratio of the mass of the charged particle to that of the neutral of 10^{-2}, and for an injection velocity equal to the mean speed. The time scale is in units of the idealized transit time for the charged particles and the electrical circuit is assumed to respond rapidly to changes of current. The current waveform shows an initial rapid decrease caused by the backscattering of a large fraction of the injected electrons to the cathode where they are absorbed. This is followed by a plateau region in which the number of charge particles in the gap is essentially constant. Finally, there is a gradual decrease in the current as the charged particles, which have been spread by diffusion, are collected by the anode. If the transit time is defined as the time at which the current reaches one half the plateau value, then this and other calculations show that the apparent drift velocity is higher than the value calculated in absence of density gradients by a factor of $(1 + 1.7\ \beta)$ for wide range of mass ratios. This compares with a factor of $(1 + 3\ \beta)$ from analysis using conventional diffusion theory [J. J. Lowke, Austral. J. Phys. 15, 39 (1962)].

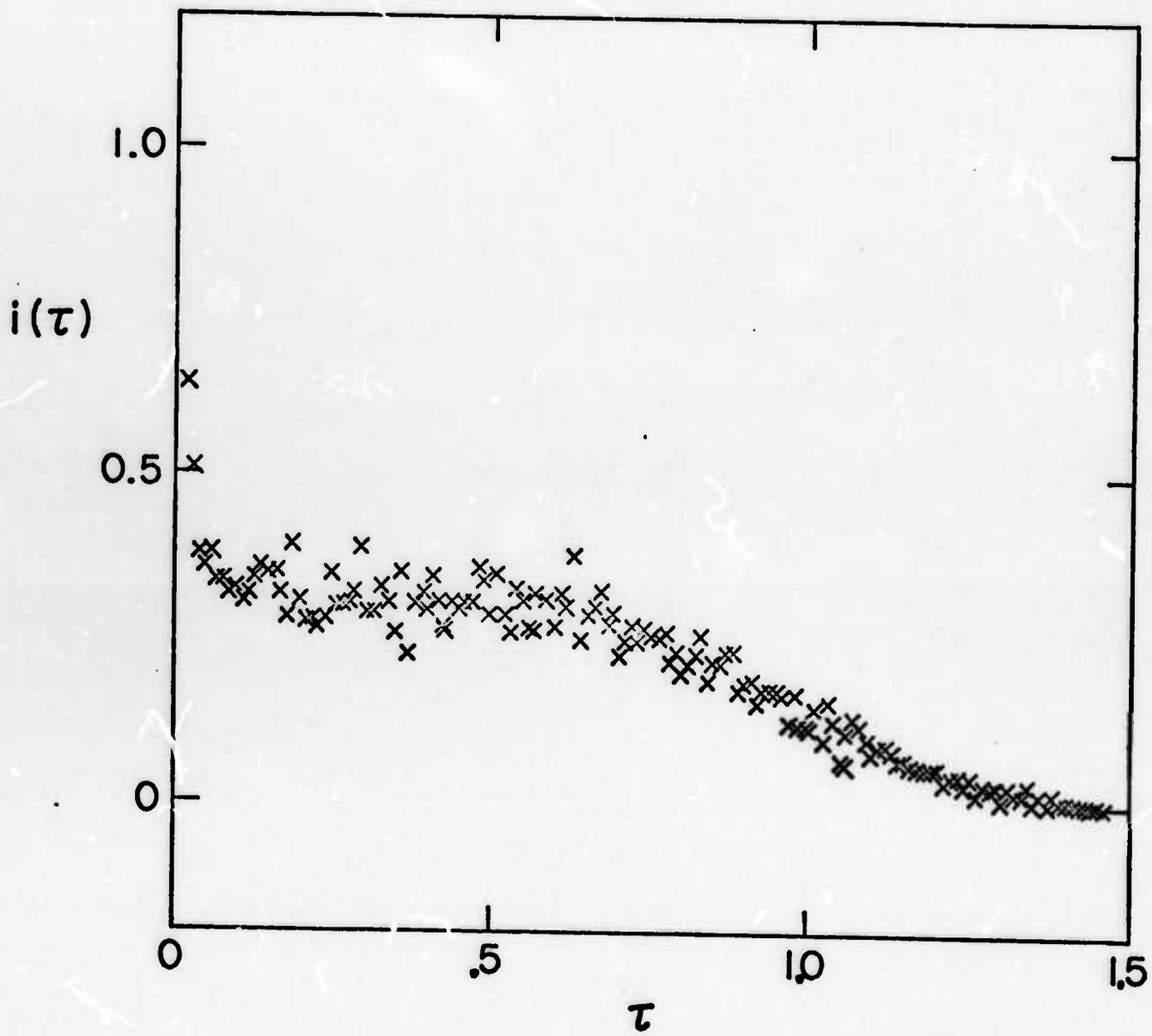

Fig. 1. Monte Carlo calculation of electron current in parallel plate drift tube used for measurement of electron drift velocity. The parameters of the calculation and interpretation of the waveform are given in the text.

3) In anticipation of the completion of the theoretical analyses of transport coefficients discussed in the preceeding subsection, we have initiated the experimental measurements of rate coefficients for the electron excitation of molecular metastables as discussed in our Management Reports and in the proposed work for FY76.

The objective of this project is the experimental determination of the rate coefficients for the production of weakly radiating states of molecules of current and potential laser interest. Our previous work in this area resulted in a) the measurement of rate coefficients for the electron excitation for the 001 level of CO_2 in pure CO_2 and in CO_2-N_2 mixtures and b) the development of apparatus for the determination of electron excitation rate coefficients for metastable states of O_2, i.e., the O_2 ($b^1\Sigma_g^+$) state, through measurement of the weak radiation emitted by the molecules. Very recently this apparatus has been used to measure the variation with O_2 density of the lifetime of $O_2(^1\Sigma_g^+)$ molecules and to obtain initial measurements of the electron excitation coefficients for this state. Figure 2 shows measured values of the reciprocal of the $O_2(^1\Sigma_g^+)$ lifetime, i.e., the decay constant, as a function of the density of oxygen atoms. At low O_2 densities our results are in agreement with earlier data [J. F. Noxon, J. Chem. Phys. 52, 1852 (1970)] and show that the loss of excited molecules is by diffusion to the cell walls. At the highest O_2 densities our decay constants are about 1/3 of Noxon's values and yield a rate coefficient of 4.5×10^{-17} cm^3 for $O_2(^1\Sigma_g^+)$ destruction by O_2 at 300°K.

The calibration of the absolute intensity scale for the measurement of the 762 nm radiation emitted by the $O_2(b^1\Sigma_g^+)$ molecules is not yet complete. However, at mean electron energies below 1.5 eV the magnitude of

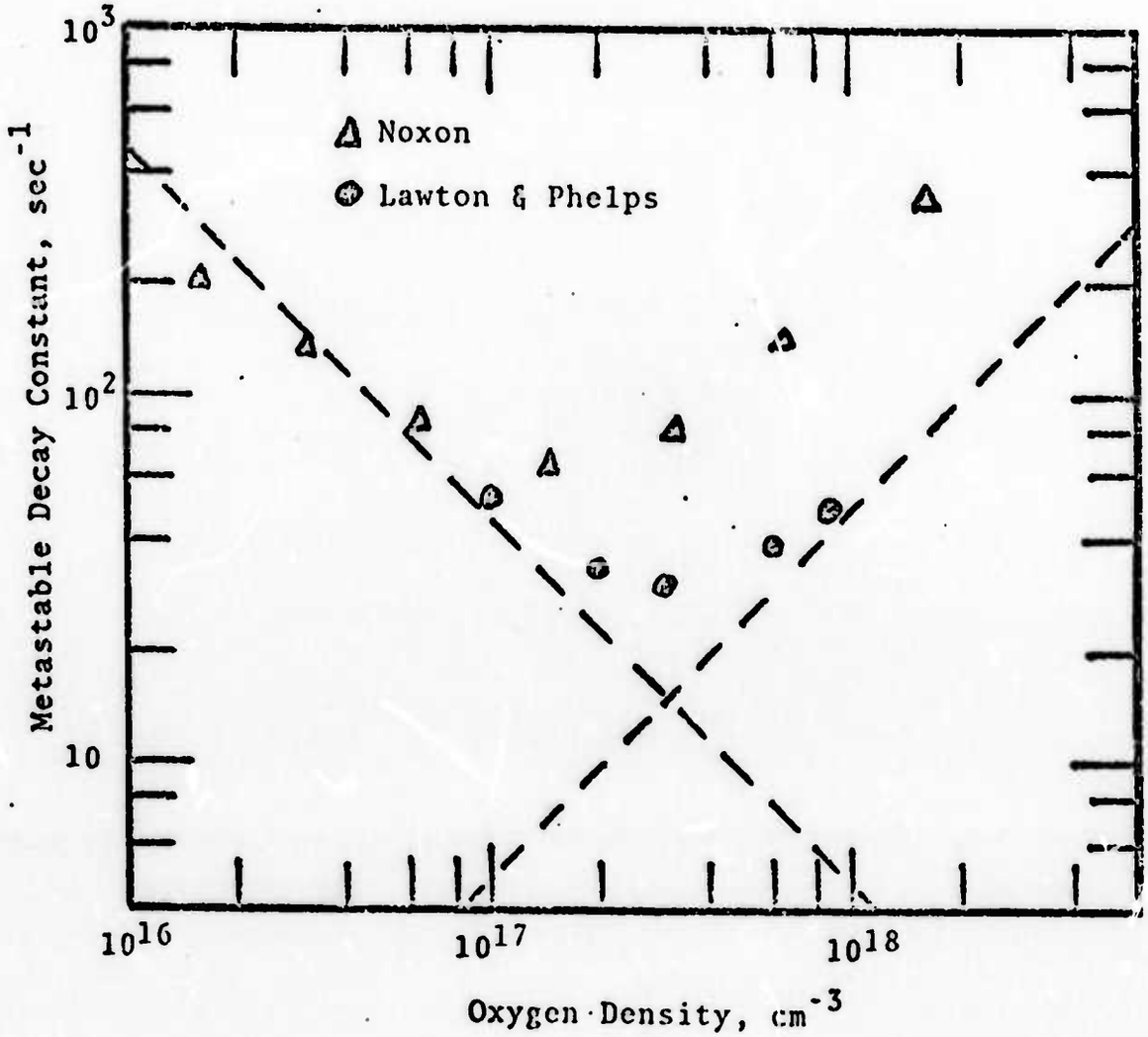

Fig. 2. Measured decay constants for the $O_2(^1\Sigma_g^+)$ states in pure O_2. The points give experimental data of Noxon and of Lawton and Phelps. The dashed lines show the asymptotic behavior expected for metastable loss by diffusion at low densities and by collisional quenching at high densities.

the rate coefficients obtained thus far are significantly below the values calculated using our recently revised set of electron excitation cross sections for O_2, including the results of electron beam measurements of the $O_2(b^1\Sigma_g^+)$ cross section [S. Trajmar, D. C. Cartwright and W. Williams, Phys. Rev. A 4, 1482 (1971) and F. Linder and H. Schmidt, Z. für Naturforsch. 26a, 1617 (1971)]. For mean electron energies greater than about 3 eV the apparent electron excitation coefficients increase with increasing mean electron energy to values almost an order of magnitude above the calculated values. We interpret this large excitation rate as being the result of dissociative excitation of O_2 to form $O(^1D)$ and the subsequent excitation transfer to form $O_2(b^1\Sigma_g^+)$.

IV. GENERATION AND INTERPRETATION OF MOLECULAR CONTINUUM RADIATION (Drs. A. C. Gallagher and Richard Scheps)

We have measured the temperature-dependent spontaneous emission spectra of Tℓ-noble gas molecules. The molecular bands on the wings of the 378 nm and 535 nm resonance lines of Tℓ have been studied, and interaction potentials for $Tℓ(6^2P_{1/2})$-Xe, $Tℓ(6^2P_{3/2})$-Xe, and $Tℓ(7^2S_{1/2})$-Xe states have been determined from the data. The Tℓ-Xe molecule is the best excimer laser candidate we have found as the fractional intensities in the far red wings of the resonance lines are largest obtained thus far. The observed spontaneous emission spectra are shown in Fig. 3. From this data we can infer the absorption and stimulated emission coefficients that would apply to a Tℓ-Xe plasma. An example is given in Fig. 4 for Tℓ and Xe densities characteristic of expected excimer lasers and for 2% of the Tℓ excited to the 7^2S state.

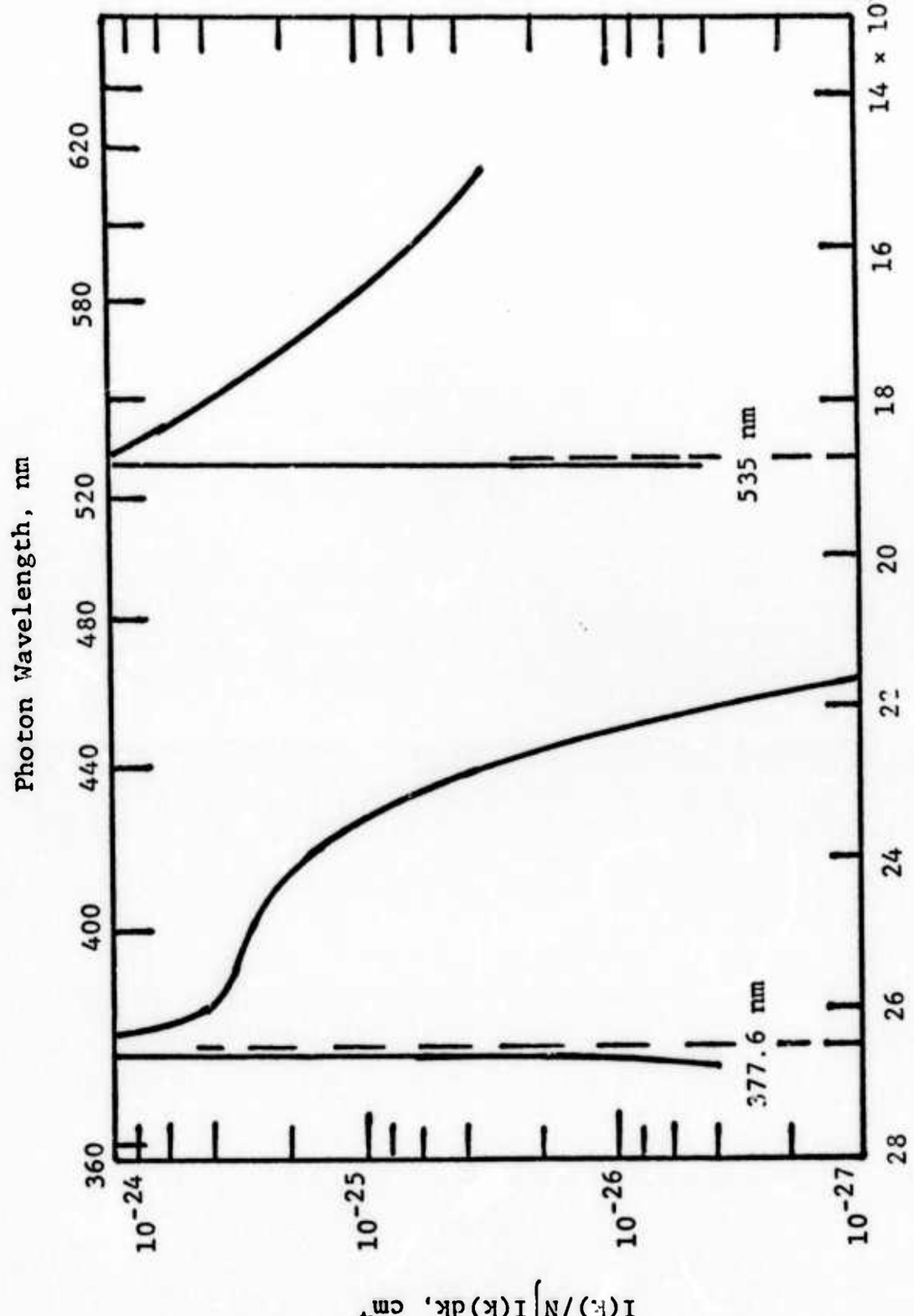

Fig. 3. Tl-Xe_2 fluorescence normalized to the Xe density N. The intensity per wave number in wing of the 3776 Å line is normalized to the total intensity in that line, and similarly for the 5350 Å line.

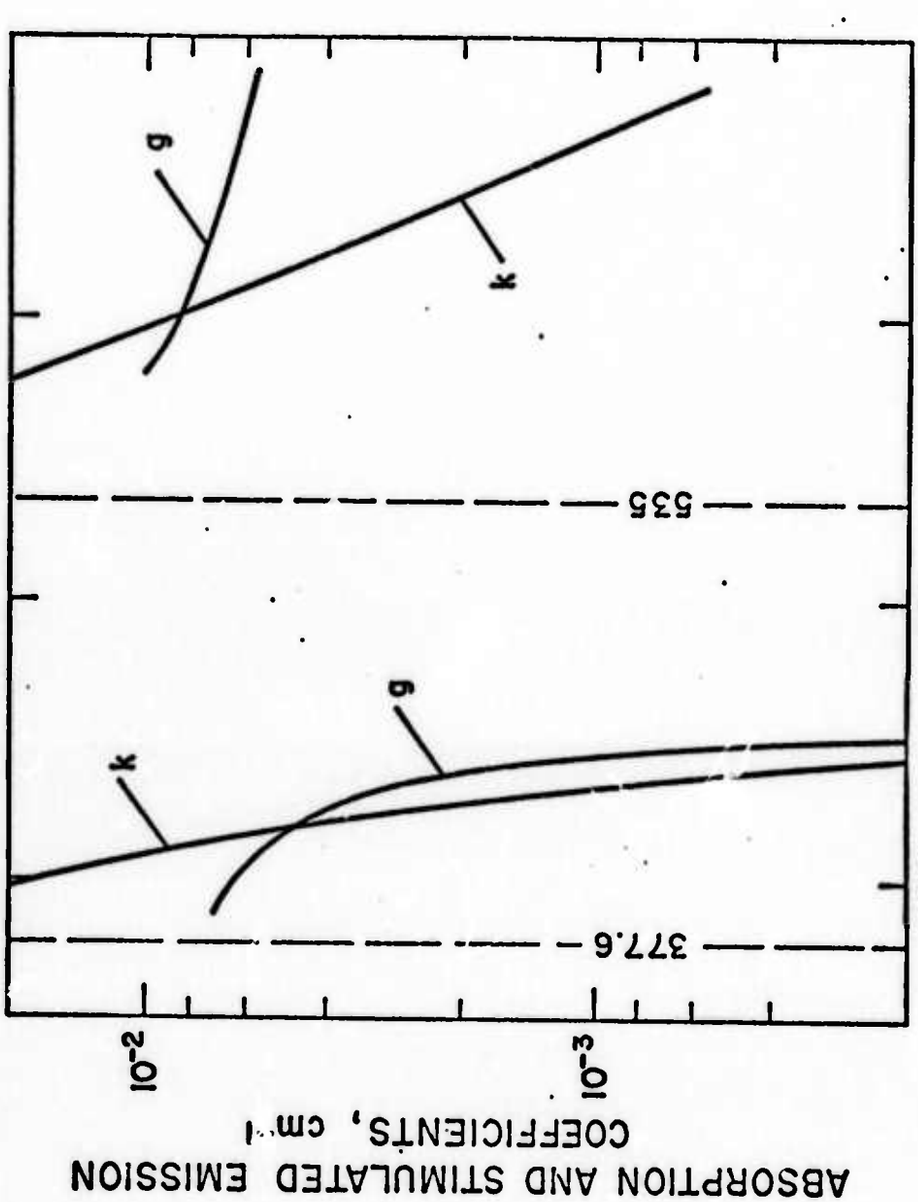

Fig. 4. Absorption (k_ν) and stimulated emission (g_ν) coefficients for Tℓ-Xe vapor, for [Xe] = 2.5 × 10^{20} cm^3 and [Tℓ] = 7.7 × 10^{16} cm^{-3}. $[Tℓ(7^2S)]/[Tℓ(6^2P_{1/2,3/2})]$ = 0.02 with equal $6^2P_{1/2}$ and $6^2P_{3/2}$ densities is also assumed.

V. SCATTERING AND TRANSPORT OF RESONANCE RADIATION IN GASES (Drs. T. Fujimoto and A. V. Phelps)

During this contract the development of apparatus and techniques for the measurement of the transport and scattering of resonance radiation in alkali metal vapors has been completed. In addition, a series of measurements have been made of the absorption, forward scattering and backward scattering of "white" light by sodium vapor for densities between 3×10^{11} and 3×10^{15} atom/cm^3 and cell thickness of 4 mm. The initial analysis of the data has been concerned with the integrated intensities or equivalent widths of the line profiles. The integrated intensities for scattering are normalized such that if all of the absorbed photons are reradiated then the sum of the equivalent widths for forward and backward scattering sum to that for absorption. Thus, Figure 5 shows the measured equivalent widths for the 589.0 nm (D_2) line of Na. The equivalent widths for the 589.6 nm (D_1) line are very similar in magnitude to that obtained by shifting the data of Fig. 5 to one half the Na densities shown. The solid lines of Fig. 5 show the calculated equivalent widths for absorption of the D2 line for temperatures of 400 and 700 K. In these calculations the splitting of the absorption lines caused by hyperfine structure has been neglected. One therefore expects agreement between measurements and theory only for a) low enough Na densities, i.e., less than about 10^{12} cm^{-3}, so that the vapor is optically thin to the resonance radiation or b) high enough densities, i.e., greater than about 10^{15} cm^{-3}, such that the measured equivalent widths are large compared to the hyperfine spacing for the D2 line. One sees from Fig. 5 that the agreement between experiment and theory is good at the low and high density limits of the present data but is poor at intermediate densities.

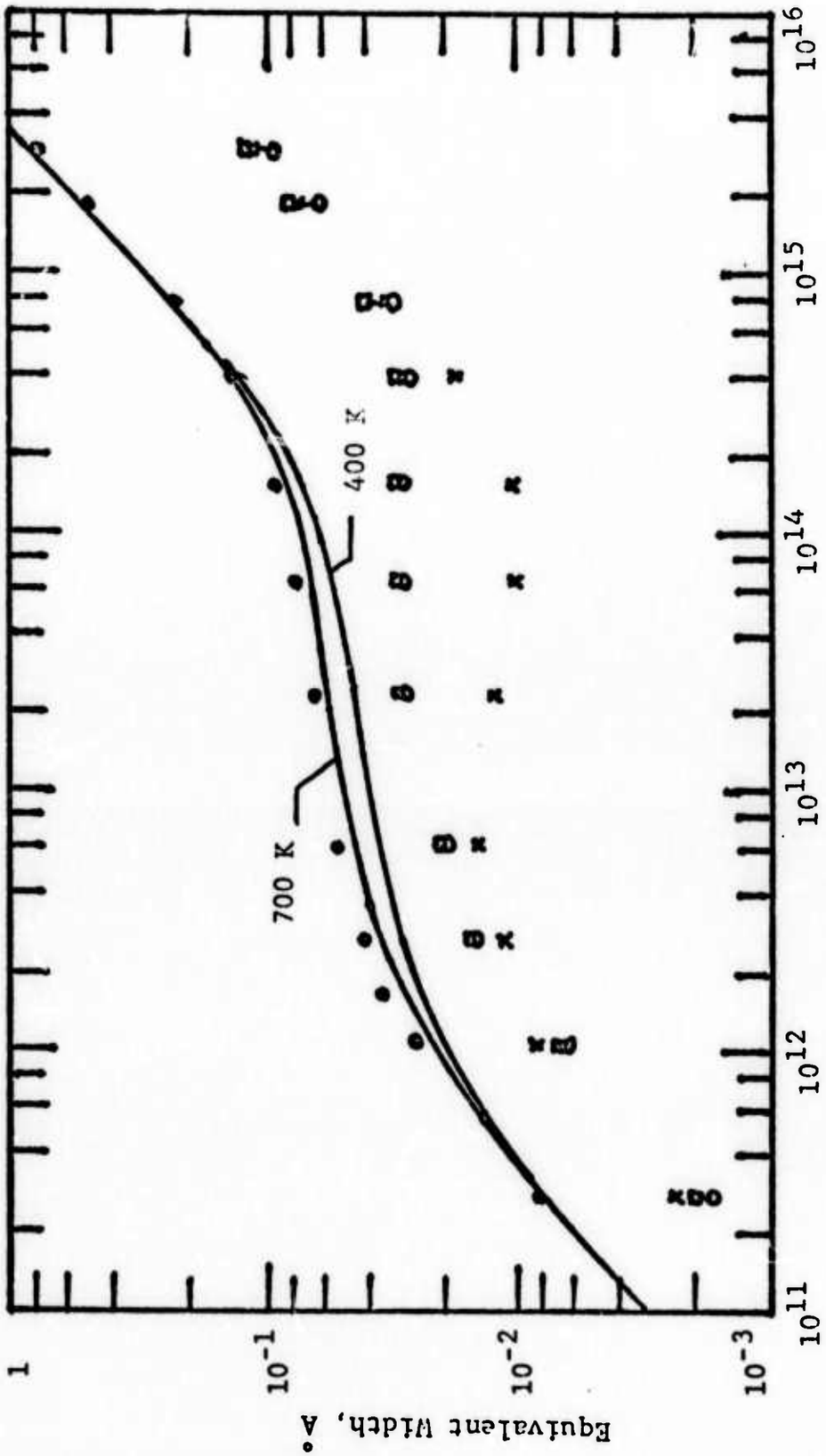

Fig. 5. Integrated absorption (●), forward scattering (X), and backward scattering (O—□) for white light illumination of Na vapor. The integrated values are expressed as equivalent widths. The solid lines are theoretical predictions of the equivalent widths neglecting hyperfine structure.

Future work will include comparisons of the integrated widths, i.e., intensities, with calculations using the detailed theory of radiation and excited atom transport developed for this project [D. G. Hummer and P. B. Kunasz, J. Quant. Spectrosc. Radiat. Trans. (in press)]. Comparisons will also be made with the simplified theories which we have developed. Such simplified theories are essential to the analyses of the electrical discharges in, for example, metal vapor rare gas systems being considered for laser applications. We also expect to obtain information regarding the collisional quenching of excited atoms at the higher vapor densities used in these experiments and in laser systems, e.g., quenching of excited Na by the few percent of Na_2 present in the vapor.

VI. PUBLICATIONS FOR FY75

Ch. Ottinger, R. Scheps, G. York, and A. Gallagher, "Broadening of the Rb resonance lines by noble gases," Phys. Rev. 11, 1815 (1975).

G. York, R. Scheps, and A. Gallagher, "Spectra and potentials of Na-noble gas molecules," J. Chem. Phys. 63, 1052 (1975).

R. Scheps, Ch. Ottinger, G. York, and A. Gallagher, "Spectra and potentials of Li-noble gas molecules," J. Chem. Phys. (in press).

E. F. Jaeger, L. Oster and A. V. Phelps, "Growth of Thermal Constrictions in a Weakly Ionized Gas Discharge: Basic Model and Numerical Solutions for Helium," Phys. Fluids (submitted).